Inelastic UV Scattering as a new Technique to Investigate Collective Excitations in Condensed Matter Physics

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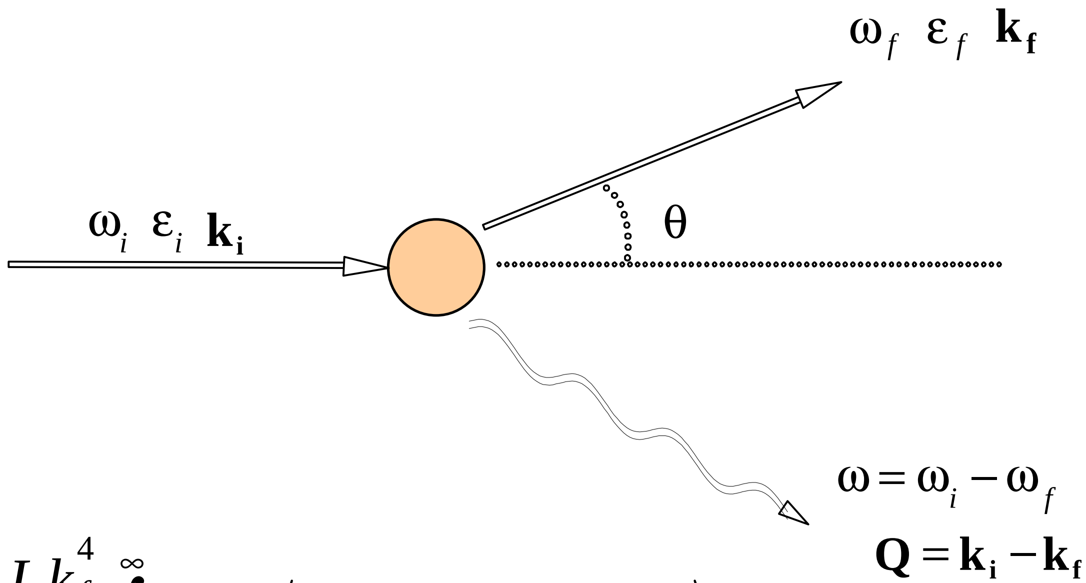
- **Inelastic UV Scattering (IUVS) with μeV Energy Resolution**
- **The Beam-line Design and Construction**
- **Applications to Condensed Matter Physics and Preliminary Results**
- **Conclusions**

Inelastic UV Scattering with μeV Energy Resolution



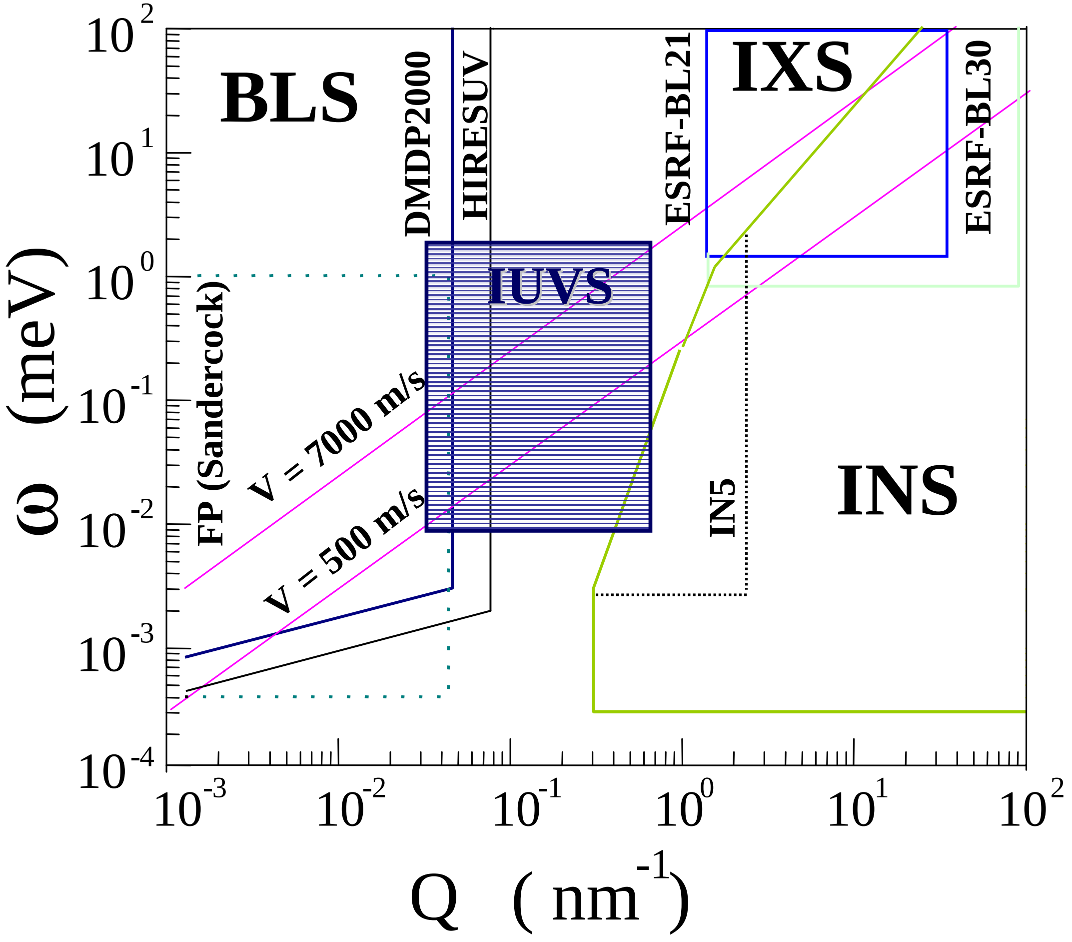
$$I_{if}(\mathbf{Q},\omega) = \frac{I_0 k_f^4}{R} \int_{-\infty}^{\infty} dt e^{-i\omega t} \langle \delta\alpha_{if}(\mathbf{Q},0)\delta\alpha_{if}(\mathbf{Q},t) \rangle$$

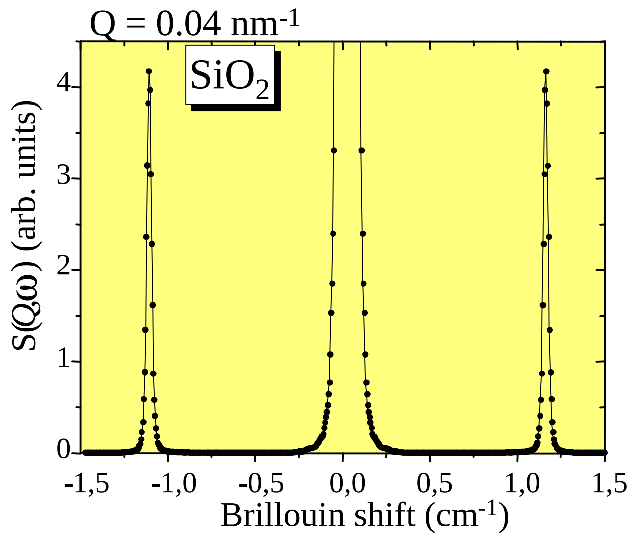
Spectral Density of Polarizability Tensor Fluctuations

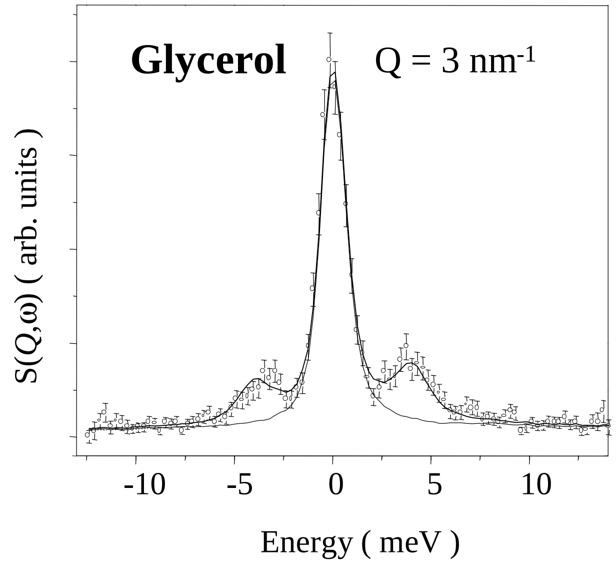
Spherical Molecules: $$I_{VV}(\mathbf{Q},\omega) = \alpha^2 S(\mathbf{Q},\omega)$$

Density Fluctuation Spectrum

Available probes to investigate collective excitations







Investigations in the Intermediate Region could Shed Light on:

Liquids - Fluids

- **Transition from the Hydrodynamic to the Kinetic regime in Simple liquids and fluids.**
- **Effect of the Local Structure on the Collective Dynamics in Molecular liquids, Associated liquids, and H-bonded liquids with a specific interest in Water and Water Solutions.**

Glasses

- **Nature of the Vibrational Modes in the Mesoscopic space-time region.**
- **Relaxational Processes in Super-Cooled liquids and their relation to the Glass Transition.**
- **Vibrational and Relaxational Low Temperature Properties of Fragile and Strong glasses.**

Resonant Scattering (Tunability)

- **Low count-rate experiments.**
- **Determination of Partial Dynamic Structure Factor.**
- **Transverse Dynamics of the system.**

Experimental requirements for IUVS

- Incident **Energy** in the 5 – 11(30) *eV* range ($\lambda \approx$ **240 – 110(40)*nm***)

- High incident photon **Flux** on the Sample (> 10^{12} *photons/s*)

  ~1 *counts/s*

- High **Resolving Power** ($\approx 10^5 - 10^6$)

$\omega \approx 10 - 10^3\ \mu eV$ $Q \approx 0.02 - 0.4\ nm^{-1}$

The Beamline Design and Construction

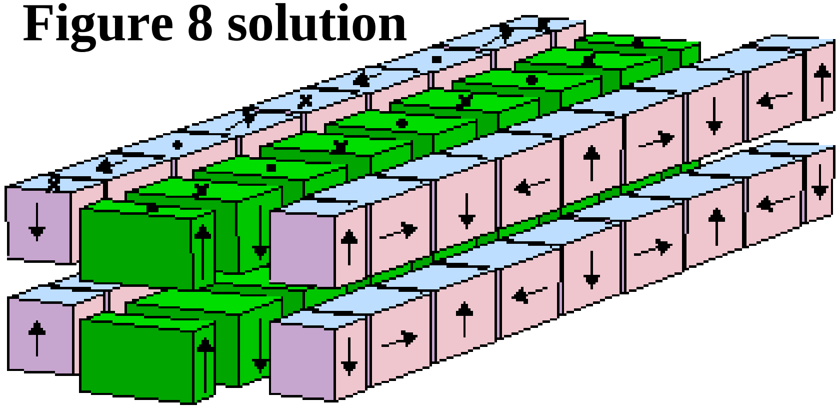
Linear Undulator ?

4.5 *m* length, 125 *mm* period, 400 *mA*

$2 \bullet 10^{15}$ *photons/s/0.1% bandwidth*

1.5 kW on the first mirror

Figure 8 solution



$N_p = 32$

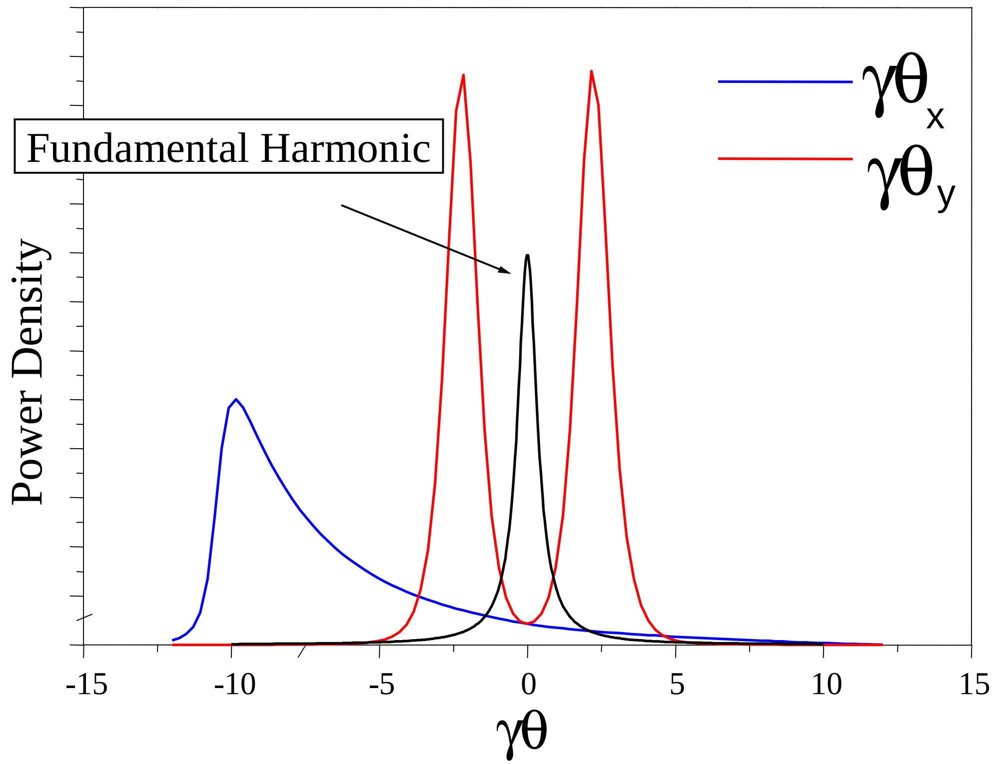
$\lambda = 140\ mm$

$K_x = 3.4$

$K_y = 9.4$

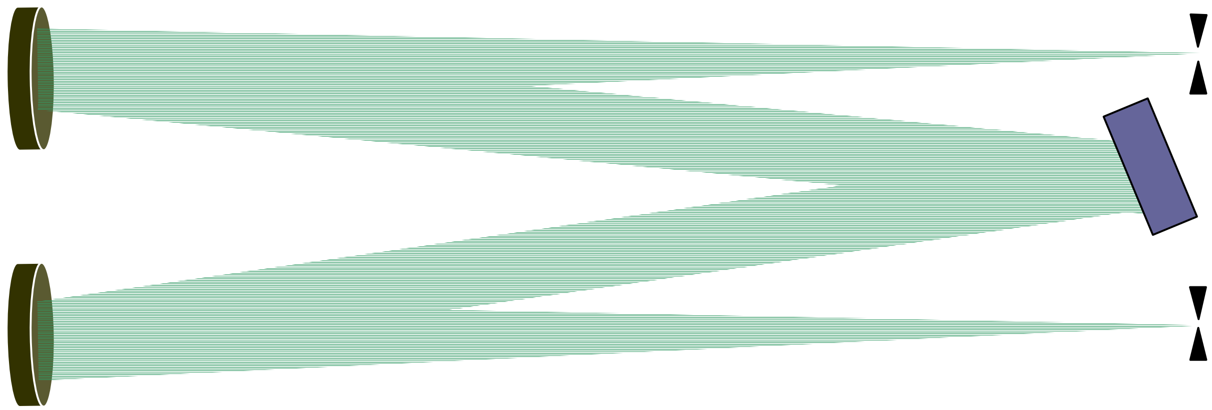
22 W on first mirror !!

$2{\cdot}10^{15}$ *photons/s/0.1% BW* ($2{\cdot}10^{12}$ *photons/s*)



The NIM Monochromator

Normal Incidence Monochromator **NIM** (*Czerny-Turner* design)



Monochromator & Analyzer design

$$\frac{\Delta E}{E} = \frac{\delta \cdot ctg\theta}{2F} = \frac{50\mu m \cdot ctg(70^{o})}{16m} \approx 1 \cdot 10^{-6}$$

The Beamline

Scanning Resolution: 80 *nrad*

Autocollimator Control: 50 *nrad*

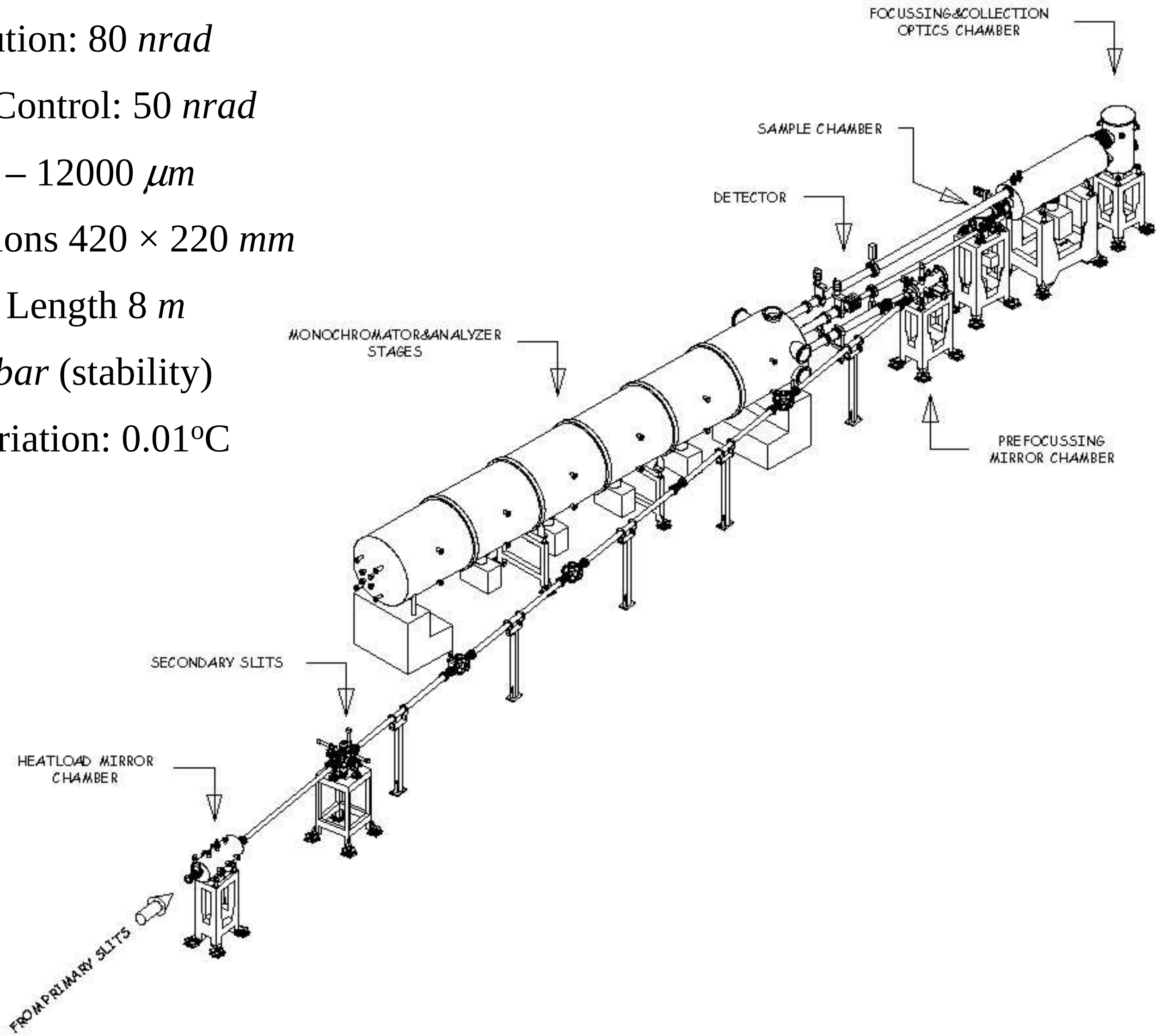
Slits Opening: 5 – 12000 μm

Grating Dimensions 420 × 220 *mm*

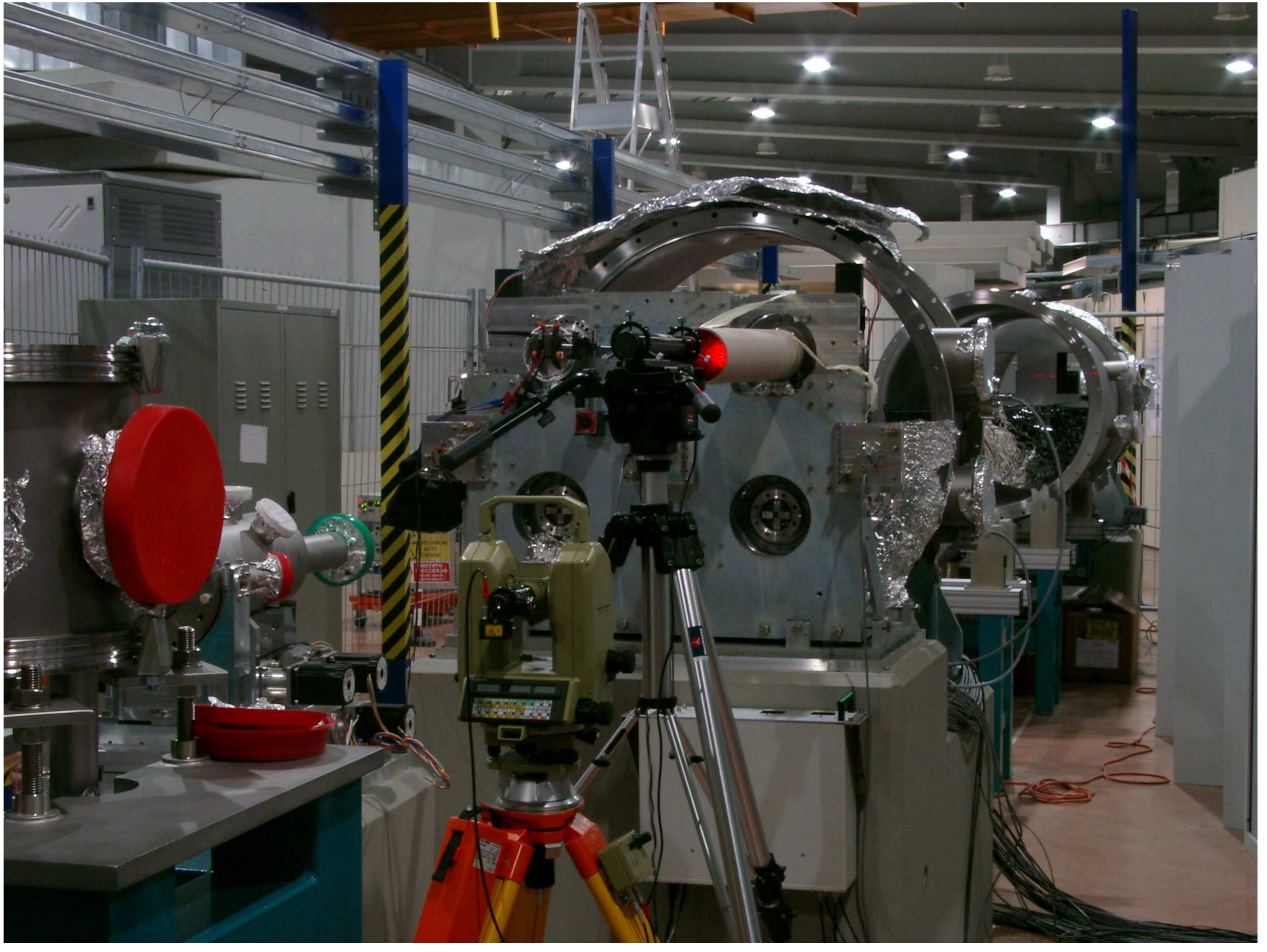
Monochromator Length 8 *m*

Vacuum: 10^{-8} *mbar* (stability)

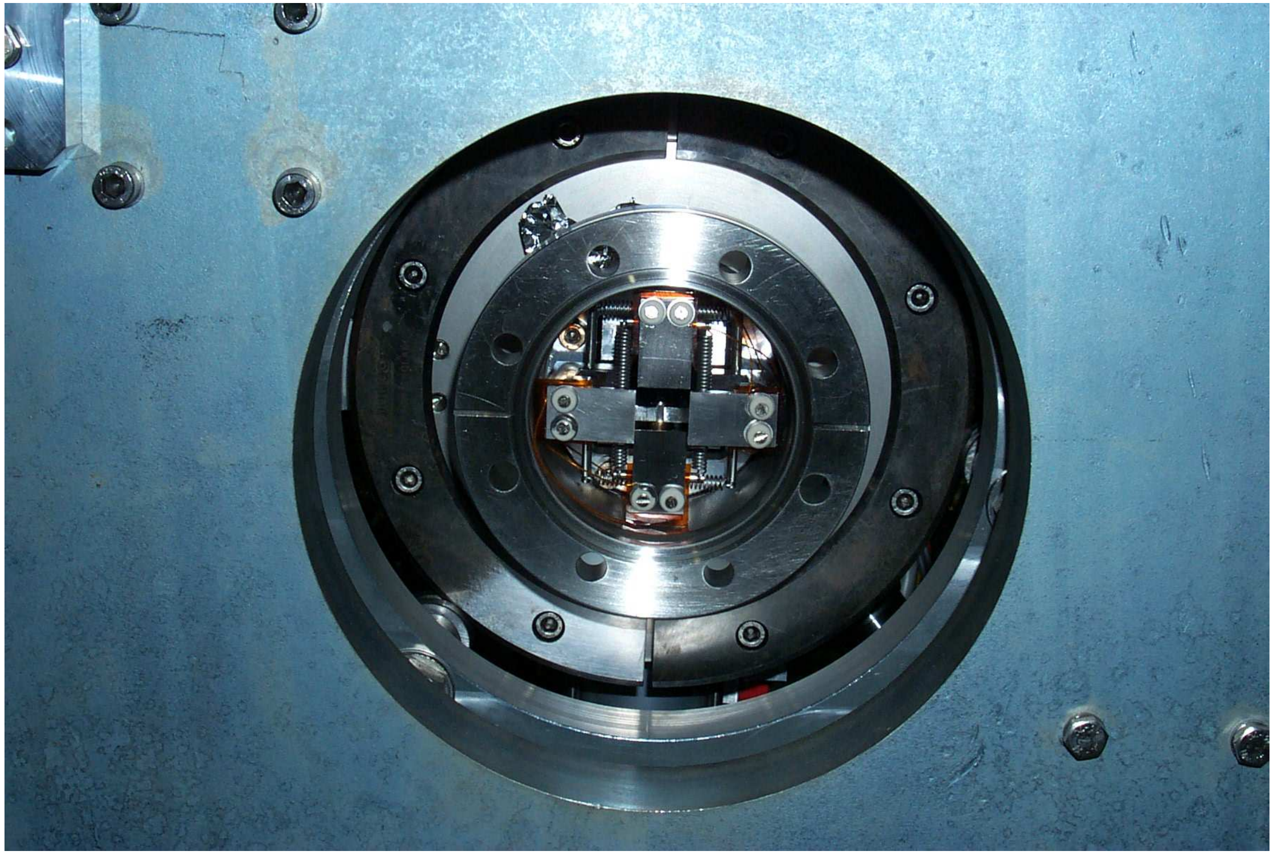
Temperature Variation: 0.01^{o}C



The Construction



The Slits

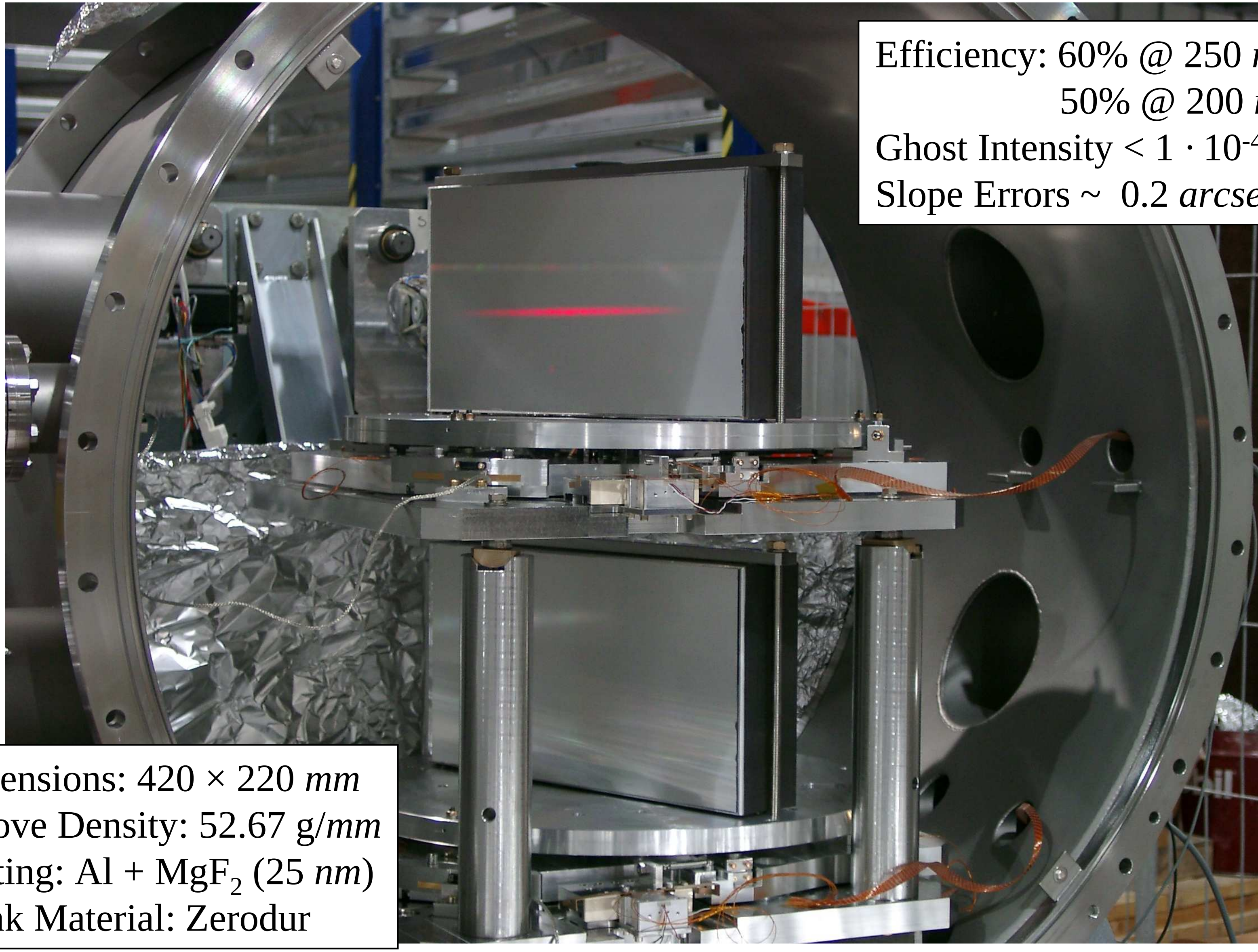


Four independent Blades design

Resolution = parallelism = repeatability : 1 μm
Rotation range (resolution): 4 *mrad* (2 μrad)
Translation range (resolution): 50 *mm* (5 μm)

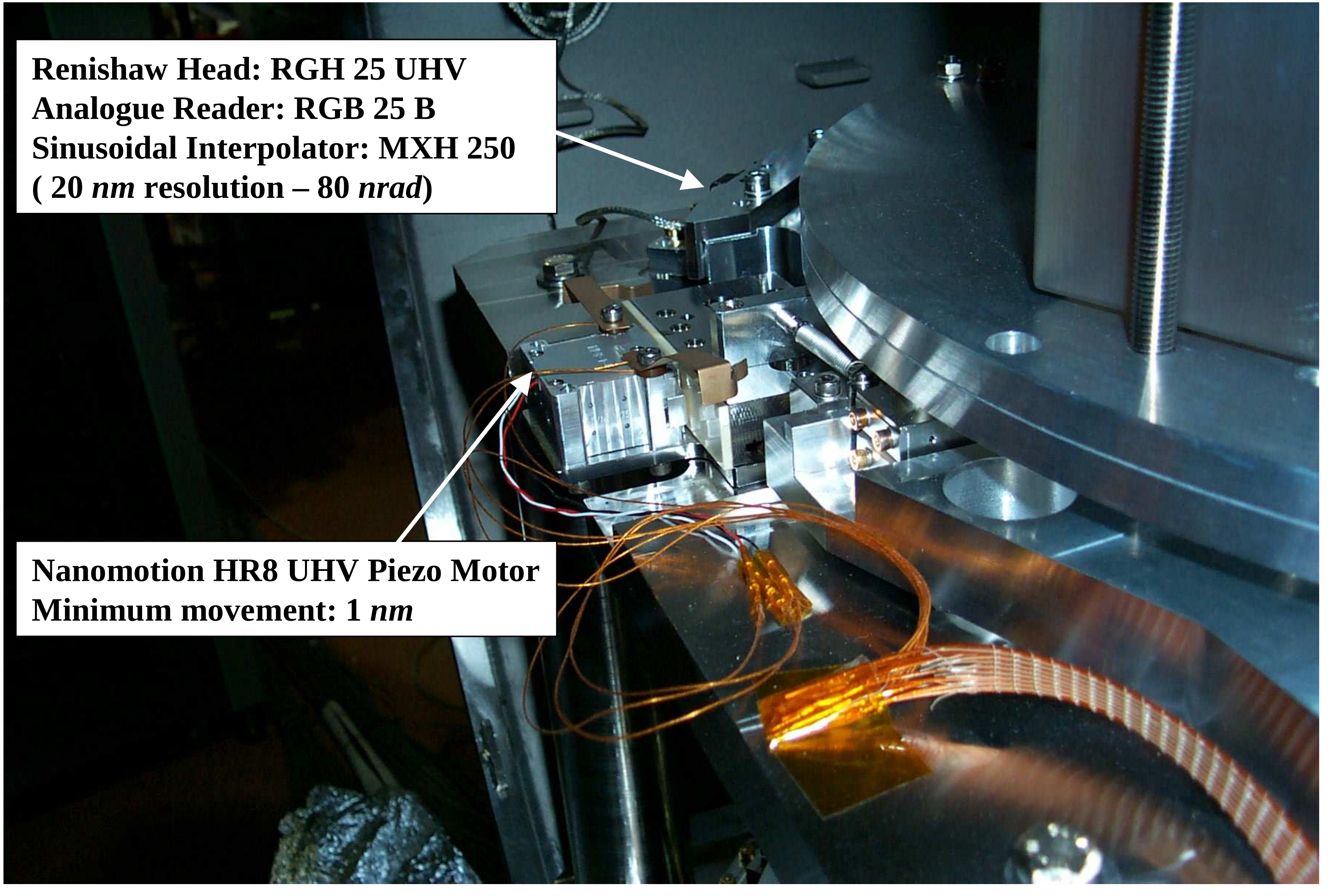
The Gratings

Efficiency: 60% @ 250 *nm*
50% @ 200 *nm*
Ghost Intensity < $1 \cdot 10^{-4}$ PL
Slope Errors ~ 0.2 *arcsec*



Dimensions: 420 × 220 *mm*
Groove Density: 52.67 g/*mm*
Coating: Al + MgF_2 (25 *nm*)
Blank Material: Zerodur

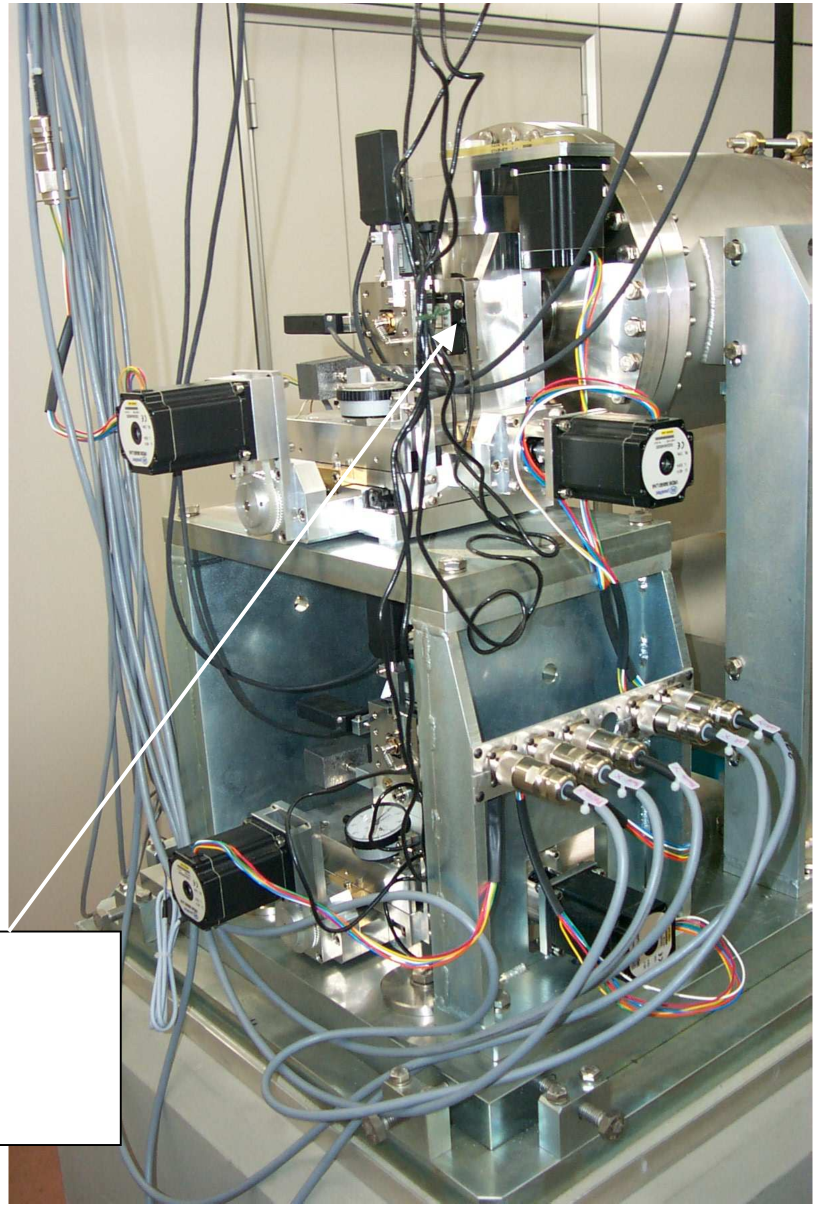
The Scanning System



Monochromator + Analyzer

Sample Stage

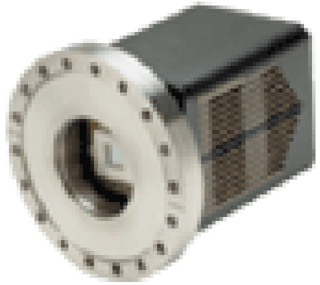
Sample Manipulator



Nanomotion Motors to achieve 1 μrad resolution movement

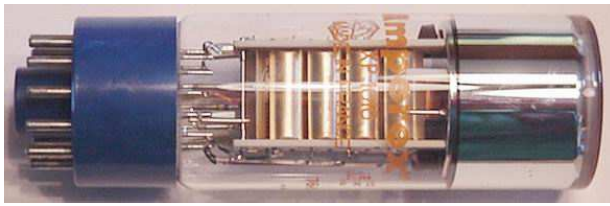
Detectors

1) **CCD** Thinned Back Illuminated
   Chip EEV 4210 13,5 μm pixel size (2048 × 515)
   Peltier cooled @ -90^{o}C ⟶ 1 *el/pixel/hour*



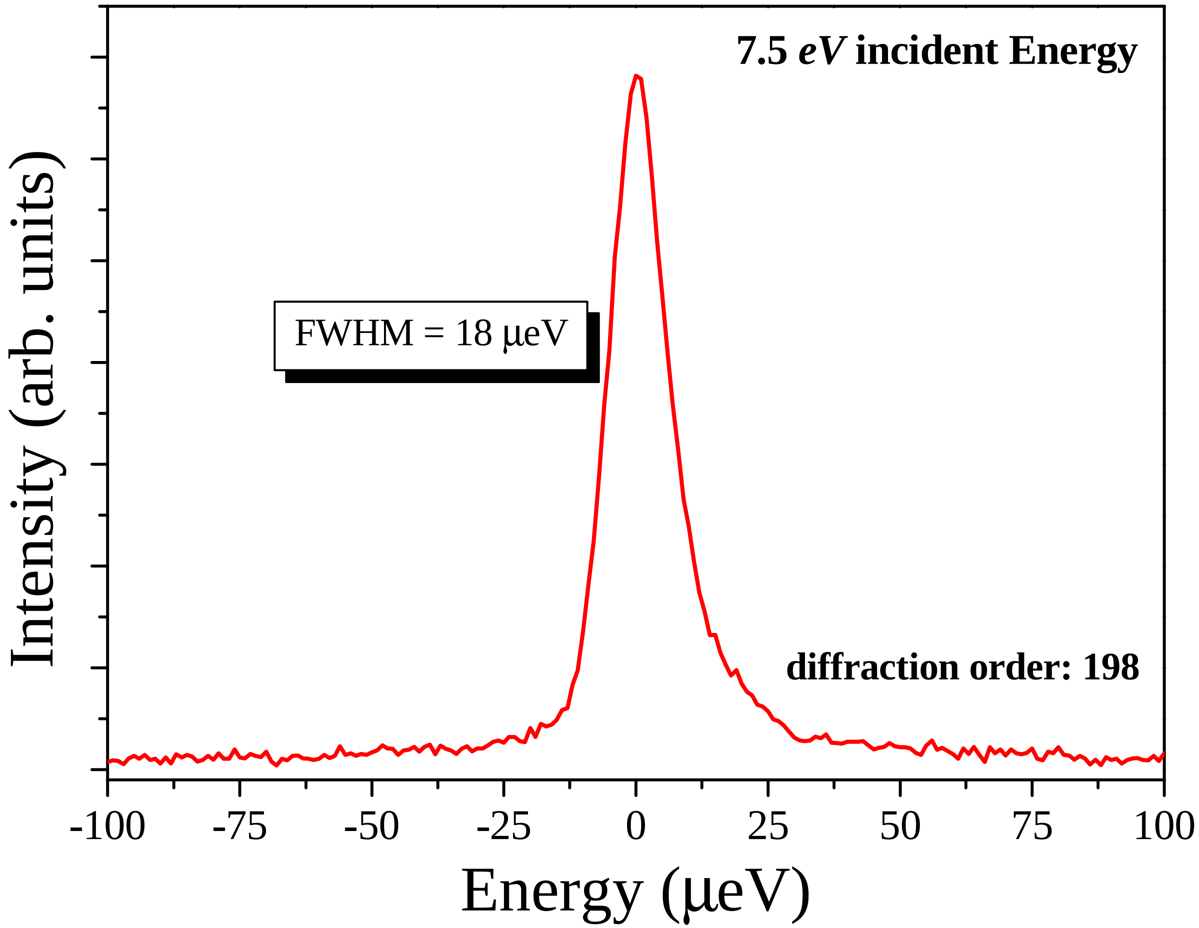
2) **MCP** coupled to a Resistive Anode
   30 μm spatial resolution
   Single Photon Counter

3) **Photomultiplier**
   no spatial resolution
   Single Photon Counter



The Resolution

Resolution measured using copper scattering



$\Delta E \approx 2 \cdot 10^{-6}$

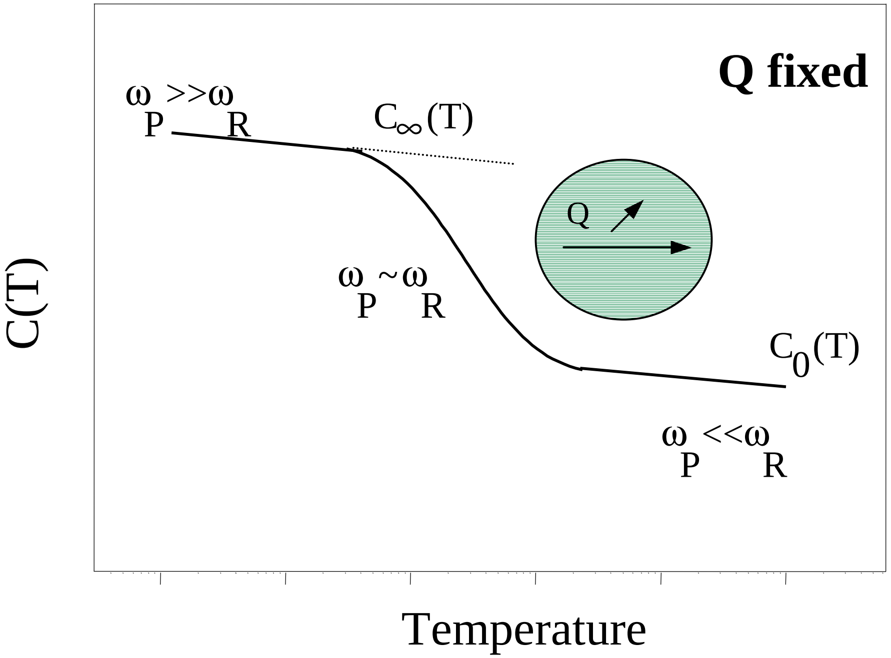
$E \approx 5 - 11$ eV

Flux $\approx 10^{12}$ ph/s

Glass-Forming Systems

Puzzling properties

- Glass-transition mechanism
- **Relaxation processes**
- Thermal anomalies
- Excess in the vibrational DOS



Knowledge of C_o, C_∞, $\tau_R(Q)$, C(Q,T)

⇓

Formulation of Models describing the Glass Transition

Structural Relaxation in Water

First evidence of glassy water: **LDA** (*Burton et al*., 1936) $T_g \approx 130\ K$

Water exhibits very unusual properties:

- Negative volume of melting
- Density maximum in the normal liquid range
- Isothermal compressibility minimum in the normal liquid range
- Increasing liquid fluidity with increasing pressure

Intermolecular forces and

Hydrogen Bond

($T \sim 4^{o}\ C$)

Evidence of a relaxation with a characteristic time of $\tau \sim 1\ ps$

high frequency investigations: **IXS** (*Sette et al.*, PRL 1996, *Monaco et al*., PRE 1999)

low frequency investigations: **Ultrasonic** (*Slie et al* ., JCP1966), **BLS** (*Cunsolo et al*., JCP1996)

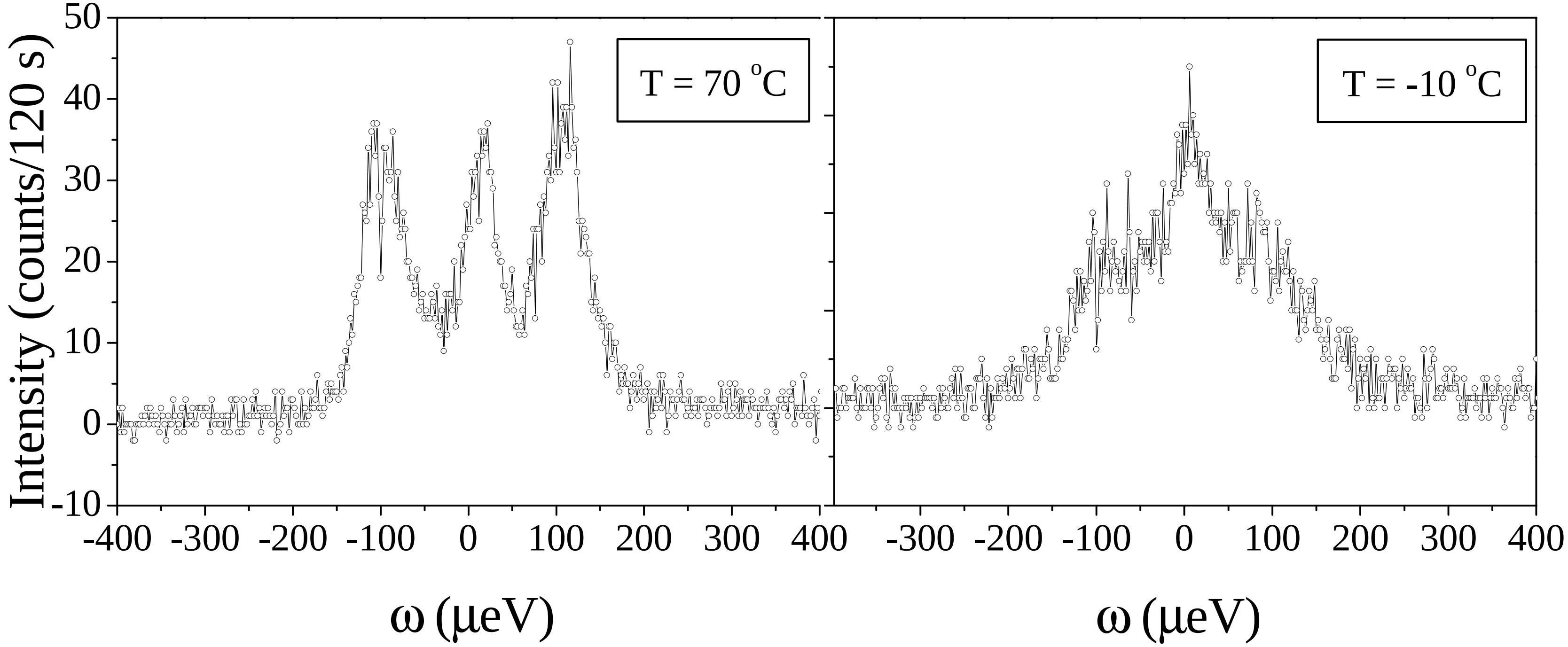
Most Sensible Region $\omega_P\ \tau_R \sim 1$ $\tau_{R\,(4^{o}C)} \sim 3\ ps$ $\omega_R \sim 200\ \mu eV$

$$\omega_P = v \cdot Q = v \cdot n \cdot 4\pi/\lambda \cdot \sin(\theta/2) \sim 100 - 250\ \mu eV$$

IUVS first measurements

Water from liquid to undercooled state

- Cell: Fused Silica Fluorescence standard Cell
- Momentum Transfer: 0.9 nm^{-1} (λ = 180 nm)
- Temperature range: -12 $\longrightarrow$ 76 ^{o}C



Conclusions

IUVS design started on March 2000. Today the beamline is in its commissioning phase and preliminary results show that it is now possible to perform inelastic scattering experiment in a Momentum-Energy transfer region not *accessible* by other experimental techniques.

The possibility to study disordered systems in the mesoscopic region will shed light on several open problems in the physics of these intriguing systems.

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